IV.F.6 Rapid-Cold Start, On-Board, Microchannel Steam Reformer

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Objectives

- Demonstrate the ability of a microchannel steam reformer and supporting heat exchangers to achieve rapid cold start.
- Develop prototype fuel processing hardware at 2-kW_e scale that will meet DOE performance targets.
- Develop reactors, vaporizers and recuperative heat exchangers that are broadly applicable to other fuel processing options.

Technical Barriers

This project addresses the following technical barriers from the Fuel Cells section of the Hydrogen, Fuel Cells and Infrastructure Technologies Program Multi-Year Research, Development and Demonstration Plan:

- I. Fuel Processor Startup/Transient Operation
- M. Fuel Processor System Integration and Efficiency

Approach

- Design and fabricate microchannel reactor, water vaporizer, air recuperator and fuel/air mixer.
- Use the low pressure drop of reactor and vaporizer to enable high startup air flows without excessive pressure drop.
- Demonstrate ability of the steam reformer and supporting heat exchangers to produce full reformate flow in <30 seconds.
- Provide recuperators, vaporizers and other microchannel components to enable other reforming research efforts.

Accomplishments

- Demonstrated that the microchannel steam reformer and supporting heat exchangers can produce full reformate flow within 12 seconds of a cold start.
- Demonstrated a warm transient of ~2 seconds.
- Fabricated a reduced-weight/higher-productivity, high-temperature, low-pressure-drop (low-dP) reforming reactor along with assembly containing supporting heat exchangers.
- Fabricated and delivered an assembly containing a high-temperature reformate recuperator and an air/reformate mixer to Argonne National Laboratory (ANL) in support of the FASTER autothermal reforming effort.
- Provided a water vaporizer to Catalytica to support their steam reformer development.

Future Directions

- Assemble and test Inconel low-dP reforming system to demonstrate
  - fast cold start and steady operation at 900°C,
  - reduction in cold-start fuel consumption,
  - bility to utilize very high-temperature combustion gas, and
  - durability of reformer to large numbers of fast-start cycles.
- Integrate reformer with water gas shift (WGS) reactor at 2-kW_e scale and demonstrate rapid cold start of the combined reformer and WGS reactor system.

Introduction

This project is applying the rapid heat and mass transfer attainable in microchannels to the development of a highly compact and efficient steam reformer. Once developed, the steam reformer will fit on-board a fuel cell vehicle and provide H_2 to a proton exchange membrane fuel cell, which in turn will produce electricity to power the vehicle. By reforming to produce hydrogen on-board the vehicle, the fuel can be gasoline, providing vehicle range, and the need to purify and compress hydrogen is eliminated. Also, an automobile with on-board reforming could be fueled using the existing gasoline infrastructure, an important advantage if a hydrogen refueling infrastructure is not available.

In past work, this project demonstrated that steam reforming, normally considered a slow reaction, can achieve rapid kinetics in a microchannel reactor. In addition, it was demonstrated that this technology could be scaled up and that it could be highly compact and efficient. In prior years, the project developed a low-pressure-drop design for a water vaporizer and demonstrated a vaporizer at 50-kW_e fuel processor scale. Recently, low-pressure-drop designs have been developed for the reforming reactor and air-air recuperator. In addition to the benefits related to reductions in parasitic power for air compression, these developments have made a rapid cold start of the steam reforming system possible. Additional information on the technology is provided in references 1, 2 and 3.

Approach

Fast-Start Reformer – The current year's work has focused on demonstrating the ability of the steam reformer and supporting heat exchangers to achieve a rapid cold start. The reformer and vaporizer have low pressure drop, enabling fast startup using high combustion gas flow. The air recuperator is bypassed during startup. The fuel is vaporized by atomizing it into steam to avoid having to heat a separate fuel vaporizer. During the startup sequence, high rates of steam are used to suppress CO generation in the reformer. This will reduce the need for WGS activity during the startup period. A higher reforming temperature system (900°C vs. 650°C) is being assembled which is expected to substantially reduce the energy needed to achieve a rapid cold start.

Recuperator/Mixer for ANL FASTER Project – The design of the low-dP air-air recuperator was adapted to the fabrication of a high-temperature recuperator for use in the ANL FASTER autothermal reformer effort.

Results

A microchannel steam reforming reactor and its associated heat exchangers demonstrated the ability to start at ambient temperature and generate full reformate flow in only 12 seconds. Microchannel components of the test system are shown in Figure 1. These components were placed within a metal duct, with the large cross section of the reforming reactor positioned across the exit of the combustion chamber. Air leaving the reforming reactor enters the water vaporizer and then exits the system, bypassing the air recuperator. Once startup is achieved, the air flow is reduced and the bypass closed to achieve high steady-state efficiency.

The reformate flow generated during a cold start is shown in Figure 2. The fuel and water pumps are started early due to a delay between the manual

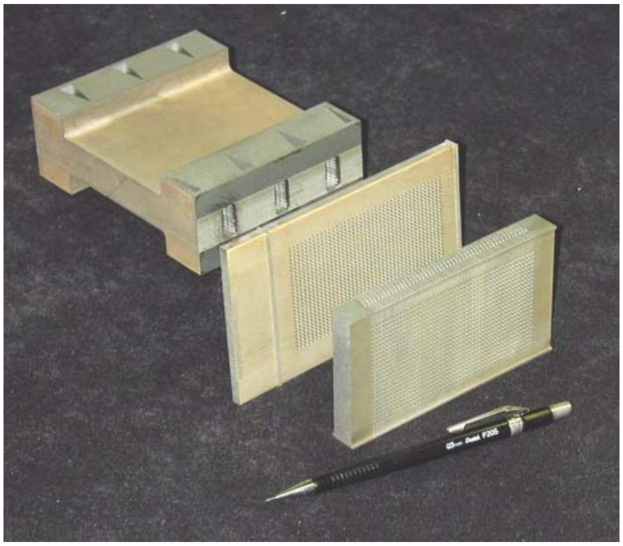

Figure 1. Key Microchannel Components of the Demonstrated Fast-Start System (From lower right to upper left are the reforming reactor, water vaporizer, and air recuperator.)

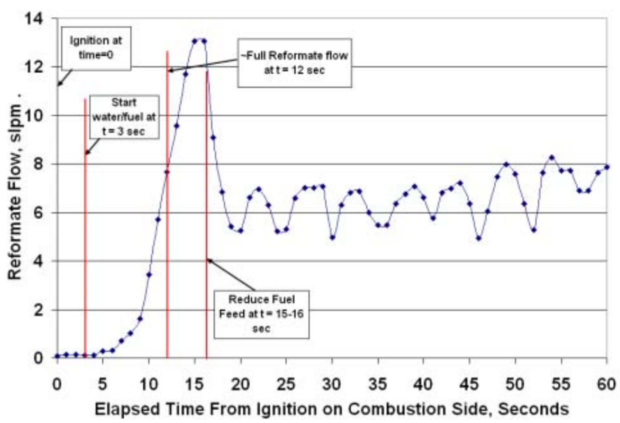

Figure 2. Measured Reformate Flow vs. Time During 12-Second Cold Start of Steam Reformer

starting of the pump and arrival of fuel and water in the system. The fuel pump was started at twice the normal flow to reduce this delay. As reformate was observed, indicating arrival of fuel, the fuel feed rate was then returned to normal. Due to the human response time, an overshoot is seen in reformate flow rate. The oscillation of the reformate rate is directly related to the output characteristic of the positive displacement pump being used to deliver the fuel. While a pulse dampener can eliminate the oscillation,

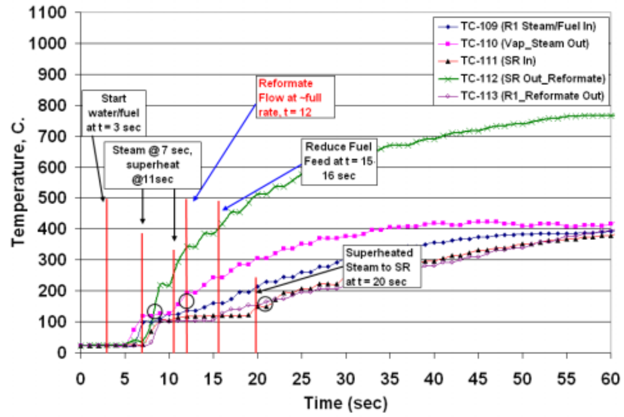

Figure 3. Reforming Side Temperatures vs. Time During 12-Second Cold Start of Steam Reformer

such a dampener might reduce transient response, so it was not included for these tests.

The system temperatures on the reformate side of the system during the fast-start test are shown in Figure 3. Several key points on the temperature profiles can be seen. First, the vaporizer outlet temperature rises to the saturation temperature within 7 seconds of the cold start. Within 11 seconds, the vaporizer temperature rises above the saturation temperature as it becomes superheated steam. This extremely fast startup of the water vaporizer is critical to the system achieving the fast start. Once superheated steam is being generated at 11 seconds, fuel can be vaporized by injecting it into the steam. At 12 seconds, full reformate flow is being produced. It is interesting to note that even though the heated reaction zone of the reformer is rapidly heated so that reforming becomes possible, the actual inlet temperature of the steam fuel mixture to the reactor does not become superheated until about 20 seconds due to the heat capacity of the connecting tubing, etc. The successful demonstration of the steam reformer starting in 12 seconds is a major step toward a complete fuel processor that can start in <1 minute.

The microchannel reformer is capable of rapid warm transient response due to the very short residence time in the system. Figure 4 shows the response observed in reformate flow to a cessation of fuel supply to the reformer. The flow of reformate drops within about 2 seconds. (The measured value does not drop to zero due to a tracer gas introduced

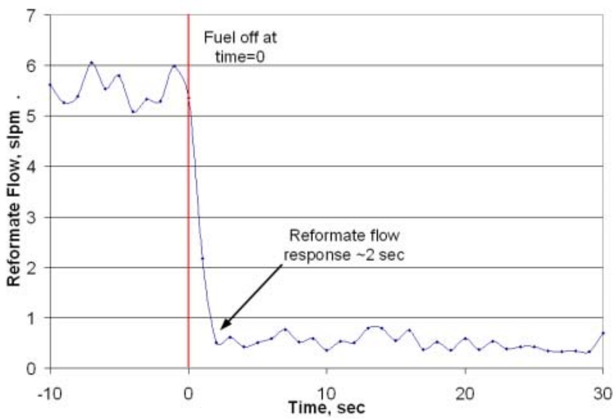

Figure 4. Rapid Warm Transient Response of Fast-Start System to Reduction in Fuel Flow

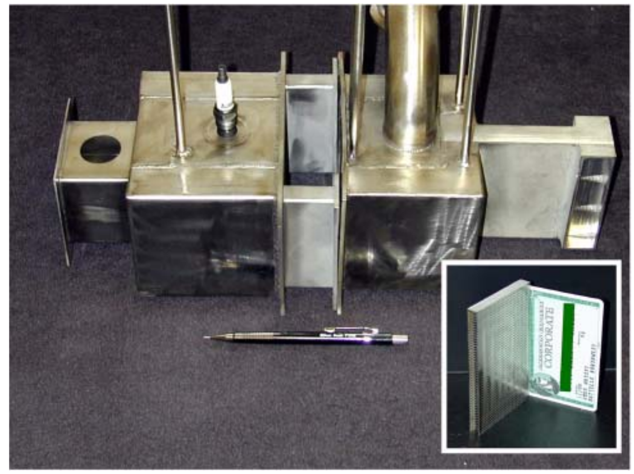

Figure 5. High-Temperature Reforming Test System and Reforming Reactor (Combustion flow is left to right. Reforming reactor (inset) and water vaporizer will be installed in the gap located in the middle of the assembly. Insulation for the combustion flow path is installed on the inside of the metal shell.)

after the reformer for analytical purposes as well as a small zero offset in the instrument.) The response is measured downstream from a condenser and a ~1 liter condensate collection vessel, which tends to dampen the response slightly. The demonstrated upward transient is ~5 seconds but is limited by the response of fuel and water delivery systems. However, there is confidence that modification of the water and fuel delivery systems would provide <2 second transients in both directions.

Now that a 12-second startup has been achieved, efforts are focusing on reducing the required air flow and fuel consumption during startup. A key part of this effort is a redesign of the reforming reactor to allow operation at higher temperature. The improved reactor has been fabricated and is shown in Figure 5. The new reactor panel is designed to operate with a reformate outlet temperature of 900°C. At this temperature, the reforming capacity of the reactor will be ~2 kW_e equivalent, roughly four times the productivity of a similar panel operating at 650°C. In addition, improvements in fabrication have achieved significant weight reduction. The new panel weighs 175 g vs. 394 g for the earlier design. This translates to a reduction in startup energy to bring the panel to operating temperature. Finally, the thickness of the panel has been reduced by ~1/3, from 0.45 inches to 0.31 inches. The reduced thickness improves the ability of the panel to tolerate very high inlet gas temperatures while controlling metal temperature via conduction between the inlet and outlet faces of the panel. This improvement will also provide a reduction in pressure drop, both during startup and steady operation.

Figure 6. Recuperator/Mixer Assembly Delivered to ANL as Part of the FASTER Project (At left the mixer assembly can be seen at the exit. Hot reformate enters the round to square transition at right.)

Finally, a microchannel recuperator and a mixer for an autothermal reformer were designed to meet ANL performance specifications, fabricated, and delivered to the ANL FASTER project. The delivered assembly, which includes the recuperator and mixer, is shown in Figure 6. The recuperator preheats incoming air and steam while cooling reformate to WGS temperatures. Key recuperator statistics are shown in Table 1. The mixer is designed to uniformly mix air into the reformate, leaving the recuperator to support the FASTER startup strategy.

Table 1. Key Metrics for Inconel 600 Microchannel Recuperator for FASTER Project

Recuperator weight (a)	1143 g
Construction	Inconel 600
Thermal duty at design point	3.6 kW
Effectiveness at design point	85%
Pressure drop, each side	~0.2 psi

(a) Weight does not include the microchannel mixer (193 g) nor tubes and ducting requested as part of the assembly.

Conclusions

The current effort has demonstrated the ability of a steam reformer with its associated heat exchangers to achieve full reformate flow within 12 seconds of a cold start. This is a major step towards a fuel processor that can start in under a minute while meeting other DOE targets. Current work is now focused on reducing energy consumption during cold startup. Table 2 provides current projections relative to DOE targets for the microchannel fuel processing system.

References

1. Whyatt, G.A., C.M. Fischer, J.M. Davis. 2002. “Development of a Rapid-Start On-Board Automotive Steam Reformer,” Conference Proceedings of 2004 AIChE Spring National Meeting, Session 12001, Innovation in Fuel Processing, April 25-29, 2004. New Orleans, LA.

2. Whyatt, G.A., C.M. Fischer, J.M. Davis. 2002. “Progress on the Development of a Microchannel Steam Reeformer for Automotive Applications,” AIChE Publication No. 164, pp 85-95. Conference proceedings, 6th International Conference on Microreaction Technology. AIChE Spring Meeting, March 10-14, 2002. New Orleans, LA.

Table 2. Development Status Compared to 2004 DOE Development Targets

Attribute	2004 Status	2004 DOE Target	Basis
Transients (sec)	~2 sec, 100%→0%	< 5 sec, 90% ↔ 10%	a
Cold Startup (20°C)	12 sec reformate, <60 sec	<60 sec, 90% traction power	b
Startup Energy (MJ/ 50 kW_e)	> 6 MJ	< 2 MJ	c
Efficiency (%)	78	78	d
Power Density (W/L)	2400	700	e
Durability (hours)	1000 hrs, 9 fast start w/o fail	2000 hrs, >50 start-stop	f
Sulfur	30 ppm spike issues, see (g)	<50 ppb from 30 ppm	g
Turndown (ratio)	10:1 with good efficiency	20:1	h

(a) Data at <2-kW_e scale. Faster response believed possible due to short residence time in hardware.
(b) Reformer data at <2-kW_e scale. Calculations based on thermal mass of WGS and preferential oxidation reactor.
(c) Estimate based on thermal mass of key reactors and heat exchangers.
(d) Earlier data at 11.2-kW_e scale with mass and energy balance calculations for low-dP system.
(e) Based on projected volume of key reactors and heat exchangers.
(f) 1000 hrs on stainless higher-dP components, fast start-stop cycles on low-dP stainless panels.
(g) Testing with 30-ppm spikes of benzothiophene and dibenzothiophene in Inconel high-temperature reactor experienced carbon deposition believed related to interaction of sulfur and Inconel surfaces.
(h) Operation of 10-20 kW_e system. Ultimately limited only by fuel pump and heat loss in WGS and preferential oxidation.

3. Whyatt, G.A., W.E. Tegrotenhuis, J.G.H. Geeting, J.M. Davis, R.S. Wegeng, and L.R. Pederson. 2001. Demonstration of Energy Efficient Steam Reforming in Microchannels for Automotive Fuel Processing. Proceedings of the Fifth International Conference on Microreaction Technology, IMRET 5, May 27-30, 2001.

FY 2004 Publications/Presentations

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